

PCT

WORLD INTELLECTUAL PROPERTY ORGANIZATION
International Bureau

D31

INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

(51) International Patent Classification ⁷ : A01N 25/08	A1	(11) International Publication Number: WO 00/47044
		(43) International Publication Date: 17 August 2000 (17.08.00)

(21) International Application Number: PCT/US00/03073	(81) Designated States: AE, AL, AM, AT, AU, AZ, BA, BB, BG, BR, BY, CA, CH, CN, CR, CU, CZ, DE, DK, DM, EE, ES, FI, GB, GD, GE, GH, GM, HR, HU, ID, IL, IN, IS, JP, KB, KG, KP, KR, KZ, LC, LK, LR, LS, LT, LU, LV, MA, MD, MG, MK, MN, MW, MX, NO, NZ, PL, PT, RO, RU, SD, SE, SG, SI, SK, SL, TJ, TM, TR, TT, TZ, UA, UG, UZ, VN, YU, ZA, ZW, ARIPO patent (GH, GM, KE, LS, MW, SD, SL, SZ, TZ, UG, ZW), Eurasian patent (AM, AZ, BY, KG, KZ, MD, RU, TJ, TM), European patent (AT, BE, CH, CY, DE, DK, ES, FI, FR, GB, GR, IE, IT, LU, MC, NL, PT, SE), OAPI patent (BF, BJ, CF, CG, CI, CM, GA, GN, GW, ML, MR, NE, SN, TD, TG).
(22) International Filing Date: 7 February 2000 (07.02.00)	
(30) Priority Data: 09/248859 11 February 1999 (11.02.99) US	
(71) Applicant: AMERICAN CYANAMID COMPANY [US/US]; Five Giralda Farms, Madison, NJ 07940 (US).	
(72) Inventors: TAKAYANAGI, Norikazu; 66-1, Nakahashira-cho, Toyohashi, Aichi (JP). KIMPARA, Masaomi; 5-5-5, Nakajima, Hamamatsu, Shizuoka (JP). SUZUKI, Munehiro; 37-8, Maseguchi, Muro-cho, Toyohashi, Aichi 441-8087 (JP).	
(74) Agents: RENDA, Barbara, L.; American Home Products Corporation, Patent Law Dept. - 2B2, One Campus Drive, Parsippany, NJ 07054 (US) et al.	
	Published With international search report.

(54) Title: LIGHT, EXTRUDED COMPOSITIONS CONTAINING A LIGHT, EXTRUDABLE, CERAMIC CARRIER, METHODS FOR THEIR USE, AND PROCESSES FOR THEIR PREPARATION

(57) Abstract

The present invention provides light, extruded compositions and methods for their preparation. The light, extruded compositions comprise at least one agricultural compound; a light, extrudable, ceramic carrier; and at least one surface active agent. The present invention also provides a method for applying agricultural compounds to the water of paddy fields by localized application(s) of the light, extruded compositions to the water surface of the paddy fields.

FOR THE PURPOSES OF INFORMATION ONLY

Codes used to identify States party to the PCT on the front pages of pamphlets publishing international applications under the PCT.

AL	Albania	ES	Spain	LS	Lesotho	SI	Slovenia
AM	Armenia	FI	Finland	LT	Lithuania	SK	Slovakia
AT	Austria	FR	France	LU	Luxembourg	SN	Senegal
AU	Australia	GA	Gabon	LV	Latvia	SZ	Swaziland
AZ	Azerbaijan	GB	United Kingdom	MC	Monaco	TD	Chad
BA	Bosnia and Herzegovina	GE	Georgia	MD	Republic of Moldova	TG	Togo
BB	Barbados	GH	Ghana	MG	Madagascar	TJ	Tajikistan
BE	Belgium	GN	Guinea	MK	The former Yugoslav Republic of Macedonia	TM	Turkmenistan
BF	Burkina Faso	GR	Greece	ML	Mali	TR	Turkey
BG	Bulgaria	HU	Hungary	MN	Mongolia	TT	Trinidad and Tobago
BJ	Benin	IE	Ireland	MR	Mauritania	UA	Ukraine
BR	Brazil	IL	Israel	MW	Malawi	UG	Uganda
BY	Belarus	IS	Iceland	MX	Mexico	US	United States of America
CA	Canada	IT	Italy	NE	Niger	UZ	Uzbekistan
CF	Central African Republic	JP	Japan	NL	Netherlands	VN	Viet Nam
CG	Congo	KE	Kenya	NO	Norway	YU	Yugoslavia
CH	Switzerland	KG	Kyrgyzstan	NZ	New Zealand	ZW	Zimbabwe
CI	Côte d'Ivoire	KP	Democratic People's Republic of Korea	PL	Poland		
CM	Cameroon	KR	Republic of Korea	PT	Portugal		
CN	China	KZ	Kazakhstan	RO	Romania		
CU	Cuba	LC	Saint Lucia	RU	Russian Federation		
CZ	Czech Republic	LI	Liechtenstein	SD	Sudan		
DE	Germany	LK	Sri Lanka	SE	Sweden		
DK	Denmark	LR	Liberia	SG	Singapore		
EE	Estonia						

LIGHT, EXTRUDED COMPOSITIONS CONTAINING A LIGHT,
EXTRUDABLE, CERAMIC CARRIER, METHODS FOR THEIR USE,
AND PROCESSES FOR THEIR PREPARATION

BACKGROUND OF THE INVENTION

Solid agricultural compositions which may be applied to the surface water of paddy fields are known in the art (see, for example, U.S. 5,833,733 and JP 6-321704).

U.S. 5,833,733 describes agrochemical formulations
5 for water surface application. These formulations
require: (1) a solid core material having an apparent
specific density of less than 1 and a particle diameter
within the range from about 300 μm to about 1,400 μm ; and
(2) at least one oily substance. However, this patent
10 does not disclose any extruded formulations. It is
difficult to prepare extruded formulations containing
large particle sizes of low-density carriers because
large, low-density particles are not entirely stable to
the conditions encountered during extrusion processes.
15 In addition, the use of oily substances is not entirely
satisfactory because oily substances may reduce the
uniformity of the extruded composition and/or may
decrease the flash point of the finished product.

JP 6-321704 describes pesticidal formulations for
20 water surface application. The formulations described in
JP 6-321704 require hollow, glass bodies having an
average particle diameter of 100 μm or less. However,
that application does not disclose any extruded
formulations. It is difficult to prepare extruded
25 formulations containing hollow, glass bodies because
hollow, glass bodies are not entirely stable to the
conditions encountered during extrusion processes.

2

It is, therefore, an object of the present invention to provide a light, extruded composition which avoids the use of low-density carriers having a particle range of about 300 μm to 1,400 μm ; oily substances; and/or hollow, glass bodies.

It is also an object of the present invention to provide a method for applying at least one agricultural compound to the water of paddy fields by applying to the water surface of the paddy fields a light, extruded composition.

It is a further object of this invention to provide a process for the preparation of a light, extruded composition.

Other objects and advantages of the present invention will be apparent to those skilled in the art from the description below and the appended claims.

SUMMARY OF THE INVENTION

The present invention relates to a light, extruded composition which comprises at least one agricultural compound; a light, extrudable, ceramic carrier; and at least one surface active agent.

The present invention also relates to a method for applying at least one agricultural compound to the water of paddy fields which method comprises applying to the water surface of the paddy fields a light, extruded composition comprising at least one agricultural compound; a light, extrudable, ceramic carrier; and at least one surface active agent.

The present invention further provides a process for the preparation of the light, extruded compositions of this invention.

DETAILED DESCRIPTION OF THE INVENTION

The light, extruded compositions of this invention comprise at least one agricultural compound; a light, extrudable, ceramic carrier; and at least one surface active agent.

Preferred light, extruded compositions of the present invention are those comprising at least one agricultural compound; a light, extrudable, ceramic carrier; at least one surface active agent; a mineral carrier; and a binder.

More preferred light, extruded compositions of the present invention are those comprising, on a weight to weight basis, about 0.5% to 75% of one or more agricultural compound(s); about 15% to 90% of a light, extrudable, ceramic carrier; about 2% to 20% of one or more surface active agent(s); about 1% to 30% of a mineral carrier; and about 0.1% to 10% of a binder.

Most preferred light, extruded compositions of this invention are those comprising, on a weight to weight basis, about 1% to 60% of one or more agricultural compound(s); about 20% to 60% of a light, extrudable, ceramic carrier; about 2% to 15% of one or more surface active agent(s); about 1% to 15% of a mineral carrier; and about 0.5% to 5% of a binder.

The light, extrudable, ceramic carrier is an especially important element of the present compositions because: (1) the light, extrudable, ceramic carrier lowers the density of the compositions, and (2) the light, extrudable, ceramic carrier does not lose its desirable low density property when subjected to the process steps used to prepare the compositions of this invention. In contrast, hollow, glass bodies, which are used in conventional light, solid compositions, are not entirely stable to conditions encountered during the preparation of light, extruded compositions. In particular, hollow, glass bodies are not entirely stable

to the pressure and shearing forces encountered in processes which include an extrusion step.

Light, extrudable, ceramic carriers suitable for use in this invention include ceramic carriers having a density less than the medium to which the light, extruded compositions are to be applied. The light, extrudable, ceramic carrier typically has a density of less than 1 g/cm³, and preferably has a density of from about 0.5 g/cm³ to 0.8 g/cm³. In a preferred embodiment of the present invention, the light, extrudable, ceramic carrier is prepared by: (1) heating a naturally occurring mineral composition which comprises silicon(IV) oxide, aluminum oxide, calcium oxide, iron(III) oxide and titanium(IV) oxide; (2) selecting particles having a density of from about 0.5 g/cm³ to 0.8 g/cm³; and (3) sieving the particles to obtain a light, extrudable, ceramic carrier having the desired particle size range. In another preferred embodiment of this invention, the light, extrudable, ceramic carrier is prepared from a composition which comprises, on a weight to weight basis, about 50% to 60% silicon(IV) oxide, about 25% to 45% aluminum oxide, about 0.1% to 6% calcium oxide, about 0.1% to 3% iron(III) oxide and about 0.1% to 2% titanium(IV) oxide. To ensure that the light, extrudable, ceramic carrier retains its desirable low density property, the light, extrudable, ceramic carrier preferably has a pressure strength (>40% survival) greater than about 150 kg/cm², and more preferably greater than about 500 kg/cm². The light, extrudable, ceramic carrier of the present invention preferably has an average particle size of about 20 μ m to 300 μ m, and more preferably, about 100 μ m to 250 μ m. Examples of light, extrudable, ceramic carriers suitable for use in the compositions of this invention include the E-SPHERES line of hollow, ceramic spheres commercially available from Taiheiyō Cement, Tokyo, Japan.

Agricultural compounds suitable for use in the light, extruded compositions of this invention include, but are not limited to, herbicides, insecticides, acaricides, nematocides, fungicides, molluscicides, plant growth regulators, safeners, algicides, and mildewicides, and mixtures thereof.

Herbicides suitable for use in the compositions of this invention include, but are not limited to, 2,4-D, 2,4-D-2-ethylhexyl, 2,4-D-isobutyl, 2,4-D-butyl, 2,4-D-calcium, 2,4-D-dimethylammonium, 2,4-D-diolamine, 2,4-D-isooctyl, 2,4-D-isopropyl, 2,4-D-potassium, 2,4-D-sodium, 2,4-D-trolamine, azimsulfuron, bensulfuron-methyl, bentazone, benzofenap, bifenox, bromobutide, butachlor, cafenstrole, chlormethoxynil, chlornitrofen, chlor-thiamid, cinmethylin, cinosulfuron, clomeprop, cyclosulfamuron, cyclosulfamuron sodium salt, cyhalofop-butyl, dimepiperate, dimethametryn, dithiopyr, dymron, esprocarb, ethoxysulfuron, etobenzanid, fentrazamid, imazosulfuron, MCPA, MCPA-butyl, MCPA-calcium, MCPA-dimethylammonium, MCPA-isooctyl, MCPA-potassium, MCPA-sodium, MCPB, MCPB-ethyl, MCPB-sodium, MCPA, mefenacet, molinate, naproanilide, oxadiazon, oxaziclomefone, pendimethalin, pentoxazone, piperophos, pretilachlor, prometryn, propanil, pyrazolate, pyrazosulfuron-ethyl, pyrazoxyfen, pyributicarb, simetryn, swep, thenylchlor, and thiobencarb, and mixtures thereof. Cyclosulfamuron, cafenstrole, and dymron, and mixtures thereof are preferred herbicides for use in the compositions of this invention.

Insecticides suitable for use in the compositions of this invention include, but are not limited to, acephate, alpha-cypermethrin, benfuracarb, bensultap, BPMC, buprofezin, carbaryl, carbofuran, carbosulfan, cartap, chlorfenapyr, chlorfenvinphos, chlorpyrifos-methyl, chlorpyrifos, cyanofenphos, cycloprothrin, diazinon, dimethoate, dioxabenzofos, disulfoton, EPN, ethofenprox, fenitrothion, fenthion, furathiocarb, isoprocarb, methyl

isothiocyanate, isoxathion, malathion, methomyl, metolcarb, monocrotophos, nitenpyram, phenthoate, pirimiphos-ethyl, pirimiphos-methyl, propaphos, propoxur, pyraclofos, pyridaphenthion, quinalphos, tetra-
5 chlorvinphos, thiocyclam, thiodicarb, trichlorphon, vamidothion, XMC, xylylcarb, and 1-[1-(p-chlorophenyl)-2-fluoro-4-(4-fluoro-3-phenoxyphenyl)-2-butenyl]-cyclopropane (R,S)-(Z), and mixtures thereof.

Fungicides suitable for use in the compositions of
10 this invention include, but are not limited to, benomyl, captan, diclomezine, edifenphos, fenoxanil, ferimzone, flutolanil, fthalide, iprobenfos, iprodione, isoprothiolane, kasugamycin, kasugamycin hydrochloride hydrate, mepronil, metalaxyl, methominostrobin, oxolinic
15 acid, pencycuron, probenazole, pyroquilon, tecloftalam, thiophanate-methyl, TPN, tricyclazole, triflumizole, and validamycin, and mixtures thereof.

Surface active agents suitable for use in the compositions of the present invention include, but are
20 not limited to, alkyl benzene sulfonates, alkyl naphthalene sulfonates, lignosulfonates, naphthalene sulfonate-formaldehyde condensates, alkyl naphthalene sulfonate-formaldehyde condensates, polyoxyethylene alkyl ethers, polyoxyethylene alkyl ether sulfates,
25 polyoxyethylene alkyl ether phosphates, polyoxyethylene alkyl aryl ethers, polyoxyethylene alkyl aryl ether sulfates, polyoxyethylene alkyl aryl ether phosphates, polycarboxylates, sodium tripolyphosphates, hexameta phosphates, phenol sulfonates, alkyl sulfates, dialkyl
30 sulfosuccinates, alkyl ether sulfates and acetylene glycols and mixtures thereof. Preferred surface active agents include alkyl naphthalene sulfonates such as a sodium alkyl naphthalene sulfonate and the like; polycarboxylates; naphthalene sulfonate-formaldehyde
35 condensates such as a sodium naphthalene sulfonate-formaldehyde condensate and the like; and dialkyl

sulfosuccinates such as a sodium alkyl sulfonated alkylate and the like; and mixtures thereof.

Mineral carriers suitable for use in the compositions of this invention include, but are not limited to, bentonites such as calcium bentonite and sodium bentonite; montmorillonite clays; kaolin clays; diatomaceous earth; and talc; and mixtures thereof. Sodium bentonite is a preferred mineral carrier.

Binders suitable for use in the compositions of the present invention include, but are not limited to, cellulose based binders such as sodium carboxymethyl cellulose, hydroxy methyl cellulose, methyl cellulose, ethyl cellulose, and the like; dextrin; starch; poly(vinyl alcohol); sodium alginate; sodium polyacrylate; and gums such as xanthan gum, welan gum, gum arabic and the like; and mixtures thereof. Sodium carboxymethyl cellulose is a preferred binder.

The present invention further provides herbicidal compositions for the control of undesirable plant species in the presence of rice plants. Preferred herbicides for the control of undesirable plant species in the presence of rice plants include cyclosulfamuron, cafenstrole, dymron, pentoxazone, fentrazamid, dithiopyr, oxaziclomefone, clomeprop, and cinmethylin, and mixtures thereof. Preferred rice herbicide mixtures for use in the compositions of this invention include: cyclosulfamuron/cafenstrole/dymron mixtures; cyclosulfamuron/pentoxazone mixtures; cyclosulfamuron/fentrazamid/dymron mixtures; cyclosulfamuron/dithiopyr mixtures; cyclosulfamuron/oxaziclomefone mixtures; cyclosulfamuron/oxaziclomefone/clomeprop mixtures; and cyclosulfamuron/cinmethylin mixtures.

A preferred herbicidal composition of this invention comprises, on a weight to weight basis, about 0.2% to 7% cyclosulfamuron; about 10% to 20% cafenstrole; about 25% to 35% dymron; about 25% to 45% of a light, extrudable, ceramic carrier which: (1) is prepared from a composition

which comprises silicon(IV) oxide, aluminum oxide, calcium oxide, iron(III) oxide and titanium(IV) oxide, (2) has an average particle size of about 100 μm to 250 μm , and (3) has a pressure strength greater than about 150 kg/cm²; about 4% to 8% of a sodium alkyl naphthalene sulfonate; about 0.5% to 2% of a mixture of a polycarboxylate and a sodium naphthalene sulfonate-formaldehyde condensate; about 1% to 3% of a sodium alkyl sulfonated alkylate; about 3% to 8% sodium bentonite; and about 1% to 3% sodium carboxymethyl cellulose.

The light, extruded compositions of this invention may be prepared by:

(a) mixing at least one agricultural compound; a light, extrudable, ceramic carrier; at least one surface active agent; optionally a mineral carrier; and optionally a binder with water to obtain an extrudable mixture;

(b) extruding the extrudable mixture from step (a) to obtain an extrudate; and

(c) drying the extrudate to obtain the light, extruded compositions of this invention.

Advantageously, the light, extrudable, ceramic carrier of the present invention is stable during the extrusion process and retains its desirable low-density property. In contrast, light, carriers made from materials such as glass and/or having a large average particle size are not entirely stable during the extrusion process and lose a significant amount of their desirable low density properties.

The light, extruded compositions of this invention may be applied directly to water, soil or other media for application of the agricultural compound(s). In particular, the light, extruded compositions of the present invention are useful for applying agricultural compounds to the water of paddy fields. Paddy-wide application of one or more agricultural compounds may be achieved by localized application(s) of the light,

extruded compositions of this invention to the water surface of paddy fields. Accordingly, the present invention provides a method for applying at least one agricultural compound to the water of paddy fields by
5 applying to the water surface of the paddy fields a light, extruded composition comprising at least one agricultural compound; a light, extrudable, ceramic carrier; and at least one surface active agent.

The light, extruded compositions of this invention
10 may be packaged in a contained, water-soluble delivery system and applied to the water surface of paddy fields in water-soluble, packaged form. Water-soluble delivery systems include, but are not limited to, water-soluble poly(vinyl alcohol) bags (PVA bags), water-soluble paper
15 bags, and water-soluble bottles, and the like.

Alternatively, the light, extruded compositions of this invention may be packaged in uncontained systems and applied by hand, spoon, aerial or mechanical application from the uncontained system. Uncontained systems
20 include, but are not limited to, bags, including foil lined bags, tubes including paper tubes, and hoppers, either hand held or attached to a mechanical applicator, such as a cultivator.

In order to facilitate a further understanding of
25 the invention, the following examples are presented primarily for the purpose of illustrating more specific details thereof. The scope of the invention should not be deemed limited by the examples, but encompasses the entire subject matter defined in the claims.

EXAMPLE 1Preparation of light, extruded compositions

- A mixture of cyclosulfamuron (3.3 g, average particle size less than 20 μm); cafenstrole (15.75 g, average particle size less than 5 μm); dymron (31.50 g); sodium bentonite (4.8 g, particle size range 5-10 μm , commercially available as bentonite-Na type from Kunimine Co., Tokyo, Japan); NEWKALGEN WG-1 a sodium alkyl naphthalene sulfonate (6.0 g, commercially available from Takemoto Oil and Fat Co., Aichi, Japan); and CELLOGEN WS-C a sodium carboxymethyl cellulose (1.5 g, commercially available from Dai-Ichi Kogyo Seiyaku Co., Kyoto, Japan) is blended to obtain a first mixture. E-SPHERES SL-180 hollow ceramic spheres (34.15 g, average particle size 115 μm , commercially available from Taiheiyo Cement, Tokyo, Japan) are added to the first mixture and blended to obtain a second mixture. NEWKALGEN TG-285 a mixture of a polycarboxylate and a sodium naphthalene sulfonate-formaldehyde condensate (1.0 g, commercially available from Takemoto Oil and Fat Co.); NEWKALGEN EP-70G a sodium alkyl sulfonated alkylate (2.0 g, commercially available from Takemoto Oil and Fat Co.); and water are added to the second mixture and blended to obtain an extrudable mixture. The extrudable mixture is passed through a basket extruder to obtain extruded material. The extruded material is dried and sieved to give the light, extruded composition identified as composition 1 in Table I.
- Using essentially the same process, the light, extruded compositions identified as compositions 2-16 in Tables I-V are obtained.

Ingredient/wt/wt										
Comp.	Cyclo- sulfamuron	Cafen- strole	Dymkon	Sodium Bentonate	NEWKALGEN WG-1	CELLOGEN WG-C	E-SPHERES/ (Avg. particle size μ m)		NEWKALGEN TG-285	NEWKALGEN EP-70G
1	3.30	15.75	31.50	4.80	6.00	1.50	34.15/ (115) ¹		1.00	2.00
2	3.30	15.75	31.50	4.80	6.00	1.50	34.15/ (103) ²		1.00	2.00
3	3.30	13.13	26.25	5.00	6.00	1.50	41.82/ (115) ¹		1.00	2.00
4	2.64	12.60	25.20	6.06	6.00	1.50	43.00/ (103) ²		1.00	2.00
5	2.20	10.50	21.00	7.30	6.00	2.00	48.00/ (103) ²		1.00	2.00

¹ E-SPHERES SL-180² E-SPHERES SL-150

TABLE II
Light, Extruded Compositions

Ingredient/wt/wt									
E-SPHERES SL-180/									
	Bensulfuron- methvl	Cafen- strole	DYMERON	Sodium Bentonite	NEWKALGEN NG-1	CELLOGE N HS-C	(Avg. particle size <u>um</u>)	NEWKALGEN TG-285	NEWKALGEN EP-70G
6	3.57	14.70	31.50	4.73	6.00	1.50	35.00/ (115)	1.00	2.00
7	2.68	11.03	23.63	5.66	6.00	2.00	46.00/ (115)	1.00	2.00
8	2.15	8.82	18.90	6.63	6.00	2.50	52.00/ (115)	1.00	2.00
9	1.79	7.35	16.75	8.11	6.00	3.00	54.00/ (115)	1.00	2.00

TABLE III
Light, Extruded Compositions

Comp.	Ingredient/wt/wt									
	Pyrazosul- furon-ethyl	Cafen- strolo	Sodium Bentonite	NEWKALGEN MG-1	CELLOGEN MS-C	(Avg. particle size μ m)	NEWKALGE N TG-285	NEWKALGE N EP-700	K-SPHERES SL-180/	
10	2.21	22.10	8.69	8.00	3.00	53.00/ (115)	1.00	2.00		
11	1.47	14.70	13.83	8.00	3.00	56.00/ (115)	1.00	2.00		
12	1.10	11.00	13.90	8.00	3.00	60.00/ (115)	1.00	2.00		

TABLE IV
Light, Extruded Compositions

Comp.	Ingredient/wt/wt									
	Pyro- Knoxanil gullon	Methomin- ostrobil	Sodium Bentonite	NEWKALGEN NG-1	CELLOGEN WS-C	(Avg. particle size μ m)	NEWKALGEN TG-285	NEWKALGEN N EP-700	X-SPHERES SL-180/	
13	50.00	-	2.00	6.00	1.00	38.00/ (115)	1.00	2.00		
14	-	50.00	2.00	6.00	1.00	38.00/ (115)	1.00	2.00		
15	-	50.00	2.00	6.00	1.00	38.00/ (115)	1.00	2.00		

TABLE V

Light, Extruded Compositions

Composition Number 16

<u>Ingredient</u>	<u>% wt/wt</u>
1-[1-(p-Chlorophenyl)-2-fluoro-4-(4-fluoro-3-phenoxyphenyl)-2-butenyl]cyclopropane (R,S)-(Z)	5.00
Carplex #80 ¹	12.00
Sodium Bentonite	10.00
NEWKALGEN WG-1	8.00
CELLOGEN WS-C	2.00
E-SPHERES SL-180 (average particle size 115 μ m)	60.00
NEWKALGEN TG-285	1.00
NEWKALGEN EP-70G	2.00

¹ A precipitated silica

EXAMPLE 2Herbicidal evaluations of light, extruded compositions

A field is cultivated, muddled, sown with weed seeds (identified below), and fertilized. Five to six days later, rice seedlings (variety Chiyonishiki) at the 2.5 leaf stage are transplanted into the prepared field. The field is then divided into 100 m² (10 m x 10 m) test plots using plastic plates and each plot is flooded with water to a depth of 5 cm. The flood levels are maintained at 5 cm throughout the evaluations. 20 g of composition number 1 from Example 1 and 20 g of composition number 1 from Example 1 packaged in water-soluble poly(vinyl alcohol) bags 20 g/bag are applied to the center of separate test plots to provide a rate equivalent to 60 g cyclosulfamuron, 300 g cafenstrole and 600 g dymron per hectare. After 42 days, the test plots are divided into quarters for herbicidal evaluations. These divisions are labeled as follows: point A - southwest quarter, point B - northwest quarter, point C - northeast quarter and point D \otimes southeast quarter. Each quarter is evaluated for percent weed control compared to a check. The results are summarized in Table VI below. As can be seen from the data in Table VI, the light, extruded composition of the present invention provides control of all weed species throughout the test plots from a single application of the invention composition in the middle of each plot.

WEED SPECIES

<u>Code Name</u>	<u>Common Name</u>	<u>Scientific Name</u>
ECHCR	Barnyardgrass	<i>Echinochloa crus-galli</i>
SCPJU	Bulrush, Japanese	<i>Scirpus juncoides</i>
MOOVA	Monochoria	<i>Monochoria vaginallis</i>
LIDPY	Falsepimpernel, common	<i>Lindernia pyxidaria</i>
SAGPY	Arrowhead	<i>Sagittaria pygmaea</i>
CYPSE	Flatsedge	<i>Cyperus serotinus</i>

TABLE VI
Herbicidal Evaluations of Light, Extruded Compositions

Treatment ¹	Weed Species	% Weed Control			
		Point-A	Point-B	Point-C	Point-D
Composition 1	ECHCR	100	100	99	100
	SCPJU	100	100	98	100
	MOOVA	100	100	100	100
	LIDPY	99	100	97	97
	SAGPY	100	100	100	100
	CYPSE	100	100	100	100
Composition 1 packaged in PVA bag	ECHCR	100	100	100	100
	SCPJU	100	100	100	100
	MOOVA	100	100	100	100
	LIDPY	100	95	100	100
	SAGPY	100	100	100	100
	CYPSE	100	100	100	100

¹ All treatments are applied at a rate to provide the equivalent of 60 g cyclosulfamuron, 300 g cafenstrole and 600 g dymron per hectare.

WHAT IS CLAIMED IS:

1. A light, extruded composition which comprises at least one agricultural compound; a light, extrudable, ceramic carrier; and at least one surface active agent.
2. The composition according to claim 1 which further comprises a mineral carrier and a binder.
3. The composition according to claim 2 which comprises on a weight basis about 0.5% to 75% of one or more agricultural compound(s); about 15% to 90% of the light, extrudable, ceramic carrier; about 2% to 20% of one or more surface active agent(s); about 1% to 30% of the mineral carrier; and about 0.1% to 10% of the binder.
4. The composition according to claim 1 wherein the light, extrudable, ceramic carrier has a density of about 0.5 g/cm³ to 0.8 g/cm³.
5. The composition according to claim 1 wherein the light, extrudable, ceramic carrier is prepared from a composition which comprises on a weight basis about 50% to 60% silicon(IV) oxide, about 25% to 45% aluminum oxide, about 0.1% to 6% calcium oxide, about 0.1% to 3% iron(III) oxide and about 0.1% to 2% titanium(IV) oxide.
6. The composition according to claim 1 wherein the light, extrudable, ceramic carrier has an average particle size of about 100 μ m to 250 μ m.

7. The composition according to claim 1 wherein the light, extrudable, ceramic carrier has a pressure strength greater than about 150 kg/cm².

8. The composition according to claim 1 wherein the agricultural compound is selected from the group consisting of a herbicide, an insecticide, an acaricide, a nematocide, a fungicide, a molluscicide, a plant growth regulator, a safener, an algicide and a mildewicide and mixtures thereof.

9. The composition according to claim 8 wherein the herbicide is selected from the group consisting of 2,4-D, 2,4-D-2-ethylhexyl, 2,4-D-isobutyl, 2,4-D-butyl, 2,4-D-calcium, 2,4-D-dimethylammonium, 2,4-D-diolamine, 2,4-D-isooctyl, 2,4-D-isopropyl, 2,4-D-potassium, 2,4-D-sodium, 2,4-D-trolamine, azimsulfuron, bensulfuron-methyl, bentazone, benzofenap, bifenox, bromobutide, butachlor, cafenstrole, chlormethoxynil, chlornitrofen, chlorthiamid, cinnmethylin, cinosulfuron, clomeprop, cyclosulfamuron, cyclosulfamuron sodium salt, cyhalofop-butyl, dimepiperate, dimethametryn, dithiopyr, dymron, esprocarb, ethoxysulfuron, etobenzanid, fentrazamid, imazosulfuron, MCPA, MCPA-butyl, MCPA-calcium, MCPA-dimethylammonium, MCPA-isooctyl, MCPA-potassium, MCPA-sodium, MCPB, MCPB-ethyl, MCPB-sodium, MCPCA, mefenacet, molinate, naproanilide, oxadiazon, oxaziclomefone, pendimethalin, pentoxazone, piperophos, pretilachlor, prometryn, propanil, pyrazolate, pyrazosulfuron-ethyl, pyrazoxyfen, pyributicarb, simetryn, swep, thenylchlor and thiobencarb and mixtures thereof; the insecticide is selected from the group consisting of acephate, alpha-cypermethrin, benfuracarb, bensultap, BPMC, buprofezin, carbaryl, carbofuran, carbosulfan, cartap, chlorfenapyr, chlorfenvinphos, chlorpyrifos-methyl, chlorpyrifos, cyanofenphos, cycloprothrin,

diazinon, dimethoate, dioxabenzofos, disulfoton, EPN, ethofenprox, fenitrothion, fenthion, furathiocarb, isoprocarb, methyl isothiocyanate, isoxathion, malathion, methomyl, metolcarb, monocrotophos, nitenpyram, phenthoate, pirimiphos-ethyl, pirimiphos-methyl, propaphos, propoxur, pyraclofos, pyridaphenthion, quinalphos, tetrachlorvinphos, thiocyclam, thiodicarb, trichlorphon, vamidothion, XMC, xylylcarb, and 1-[1-(p-chlorophenyl)-2-fluoro-4-(4-fluoro-3-phenoxyphenyl)-2-butenyl]cyclopropane (R,S)-(Z), and mixtures thereof; and the fungicide is selected from the group consisting of benomyl, captan, diclomezine, edifenphos, fenoxanil, ferimzone, flutolanil, thalide, iprobenfos, iprodione, isoprothiolane, kasugamycin, kasugamycin hydrochloride hydrate, mepronil, metalaxyl, methominostrobin, oxolinic acid, pencycuron, probenazole, pyroquilon, tecloftalam, thiophanate-methyl, TPN, tricyclazole, triflumizole and validamycin and mixtures thereof.

10. The composition according to claim 1 wherein the agricultural compound is selected from the group consisting of cyclosulfamuron, cafenstrole and dymron and mixtures thereof.

11. The composition according to claim 1 wherein the surface active agent is selected from the group consisting of an alkyl benzene sulfonate, an alkyl naphthalene sulfonate, a lignosulfonate, a naphthalene sulfonate-formaldehyde condensate, an alkyl naphthalene sulfonate-formaldehyde condensate, a polyoxyethylene alkyl ether, a polyoxyethylene alkyl ether sulfate, a polyoxyethylene alkyl ether phosphate, a polyoxyethylene alkyl aryl ether, a polyoxyethylene alkyl aryl ether sulfate, a polyoxyethylene alkyl aryl ether phosphate, a polycarboxylate, a sodium tripolyphosphate, a hexameta

phosphate, a phenol sulfonate, an alkyl sulfate, a dialkyl sulfosuccinate, an alkyl ether sulfate and an acetylene glycol and mixtures thereof.

12. The composition according to claim 11 wherein the surface active agent is selected from the group consisting of an alkyl naphthalene sulfonate, a polycarboxylate, a naphthalene sulfonate-formaldehyde condensate and a dialkyl sulfosuccinate and mixtures thereof.

13. The composition according to claim 12 wherein the surface active agent comprises a mixture of a sodium alkyl naphthalene sulfonate, a polycarboxylate, a sodium naphthalene sulfonate-formaldehyde condensate and a sodium alkyl sulfonated alkylate.

14. The composition according to claim 2 wherein the mineral carrier is selected from the group consisting of calcium bentonite, sodium bentonite, montmorillonite, kaolin clay, pyrophyllite clay, sericite clay, diatomaceous earth and talc and mixtures thereof.

15. The composition according to claim 14 wherein the mineral carrier is sodium bentonite.

16. The composition according to claim 2 wherein the binder is selected from the group consisting of sodium carboxymethyl cellulose, hydroxy methyl cellulose, methyl cellulose, ethyl cellulose, dextrin, starch, poly(vinyl alcohol), sodium alginate, sodium polyacrylate, xanthan gum, welan gum and gum arabic and mixtures thereof.

17. The composition according to claim 16 wherein the binder is sodium carboxymethyl cellulose.

18. The composition according to claim 2 which comprises on a weight basis about 0.2% to 7% cyclosulfamuron; about 10% to 20% cafenstrole; about 25% to 35% dymron; about 25% to 45% of a light, extrudable, ceramic carrier which: (1) is prepared from a composition which comprises on a weight basis about 50% to 60% silicon(IV) oxide, about 25% to 45% aluminum oxide, about 0.1% to 6% calcium oxide, about 0.1% to 3% iron(III) oxide and about 0.1% to 2% titanium(IV) oxide, (2) has an average particle size of about 100 μm to 250 μm , and (3) has a pressure strength greater than about 150 kg/cm^2 ; about 4% to 8% of a sodium alkyl naphthalene sulfonate; about 0.5% to 2% of a mixture of a polycarboxylate and a sodium naphthalene sulfonate-formaldehyde condensate; about 1% to 3% of a sodium alkyl sulfonated alkylate; about 3% to 8% sodium bentonite; and about 1% to 3% sodium carboxymethyl cellulose.

19. A method for applying at least one agricultural compound to the water of paddy fields which comprises applying to the water surface of the paddy fields a light, extruded composition comprising at least one agricultural compound; a light, extrudable, ceramic carrier; and at least one surface active agent.

20. The method according to claim 19 wherein the light, extruded composition further comprises a mineral carrier and a binder.

21. The composition according to claim 19 wherein the light, extrudable, ceramic carrier has a density of about 0.5 g/cm^3 to 0.8 g/cm^3 .

22. The method according to claim 19 wherein the light, extrudable, ceramic carrier is prepared from a composition which comprises on a weight basis about 50% to 60% silicon(IV) oxide, about 25% to 45% aluminum oxide, about 0.1% to 6% calcium oxide, about 0.1% to 3% iron(III) oxide and about 0.1% to 2% titanium(IV) oxide.

23. The method according to claim 19 wherein the light, extrudable, ceramic carrier has an average particle size of about 100 μm to 250 μm .

24. The method according to claim 19 wherein the light, extrudable, ceramic carrier has a pressure strength greater than about 150 kg/cm^2 .

25. The method according to claim 19 wherein the agricultural compound is selected from the group consisting of a herbicide, an insecticide, an acaricide, a nematocide, a fungicide, a molluscicide, a plant growth regulator, a safener, an algicide and a mildewicide and mixtures thereof.

26. The method according to claim 19 wherein the agricultural compound is selected from the group consisting of cyclosulfamuron, cafenstrole and dymron and mixtures thereof.

27. The method according to claim 20 wherein the surface active agent comprises a mixture of a sodium alkyl naphthalene sulfonate, a polycarboxylate, a sodium naphthalene sulfonate-formaldehyde condensate and a sodium alkyl sulfonated alkylate; the mineral carrier is sodium bentonite; and the binder is sodium carboxymethyl cellulose.

28. The method according to claim 19 wherein the light, extruded composition is packaged in a contained, water-soluble delivery system and the packaged, light, extruded composition is applied to the water surface.

29. The method according to claim 28 wherein the contained, water-soluble delivery system is selected from the group consisting of a water-soluble poly(vinyl alcohol) bag, a water-soluble paper bag and a water-soluble bottle.

30. The method according to claim 19 wherein the light, extruded composition is packaged in an uncontained system and the uncontained, light, extruded composition is applied to the water surface by hand, spoon, aerial or mechanical application.

31. The method according to claim 30 wherein the uncontained system is selected from the group consisting of a bag, a tube and a hopper.

32. A process for the preparation of a light, extruded composition comprising at least one agricultural compound; a light, extrudable, ceramic carrier; and at least one surface active agent, which process comprises the steps of:

(a) mixing the agricultural compound; the light, extrudable, ceramic carrier; and the surface active agent with water to obtain an extrudable mixture;

(b) extruding the extrudable mixture from step (a) to obtain an extrudate; and

(c) drying the extrudate to obtain the light, extruded composition.

. 33. The process according to claim 32 wherein step
(a) further comprises a mineral carrier and a binder.

INTERNATIONAL SEARCH REPORT

Int'l. Application No
PCT/US 00/03073

A. CLASSIFICATION OF SUBJECT MATTER
IPC 7 A01N25/08

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)
IPC 7 A01N

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	US 5 833 733 A (WADA YUZURU ET AL) 10 November 1998 (1998-11-10) cited in the application the whole document	1-33
A	DATABASE WPI Week 199506 Derwent Publications Ltd., London, GB; AN 1995-041123 XP002137030 & JP 06 321704 A (HOKKO CHEM INC CO LTD), 22 November 1994 (1994-11-22) cited in the application abstract	1-33

☒ Further documents are listed in the continuation of box C.

☒ Patent family members are listed in annex.

* Special categories of cited documents:

"A" document defining the general state of the art which is not considered to be of particular relevance

"E" earlier document but published on or after the international filing date

"L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)

"O" document referring to an oral disclosure, use, exhibition or other means

"P" document published prior to the international filing date but later than the priority date claimed

"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention

"X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone

"Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art.

"A" document member of the same patent family

Date of the actual completion of the international search

5 May 2000

Date of mailing of the international search report

23/05/2000

Name and mailing address of the ISA

European Patent Office, P.O. 5018 Patentkanal 2
NL - 2280 HV Rijswijk
Tel. (+31-70) 340-2040, Tx. 31 651 epo nl,
Fax: (+31-70) 340-3016

Authorized officer

Bertrand, F

INTERNATIONAL SEARCH REPORT

Int'l. Jonal Application No

PCT/US 00/03073

C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	WO 93 25074 A (DU PONT ;FREEMAN ROY QUINN III (US); SANDELL LIONEL SAMUEL (US); Z) 23 December 1993 (1993-12-23) page 2 -page 4 page 7, paragraph 2 page 15, paragraph 3	1-33
X	WO 95 08265 A (DU PONT ;SANDELL LIONEL SAMUEL (US); WYSONG ROBERT DAVID (US)) 30 March 1995 (1995-03-30) page 4, paragraph 3 page 6 -page 8	1-33
X	EP 0 280 289 A (TAKEDA CHEMICAL INDUSTRIES LTD) 31 August 1988 (1988-08-31) page 2, paragraph 1 page 3, paragraphs 2,5-7,10 examples 10,11	1-33

INTERNATIONAL SEARCH REPORT

Information on patent family members

Int. National Application No

PCT/US 00/03073

Patent document cited in search report	Publication date	Patent family member(s)	Publication date
US 5833733 A	10-11-1998	CN 1145719 A	26-03-1997
		ES 2119683 A	01-10-1998
		IT MI960917 A	10-11-1997
		JP 9183701 A	15-07-1997
JP 6321704 A	22-11-1994	NONE	
WO 9325074 A	23-12-1993	AU 4528693 A	04-01-1994
		BR 9306686 A	08-12-1998
		CA 2137773 A	23-12-1993
		CN 1082814 A	02-03-1994
		EP 0645961 A	05-04-1995
		HU 70290 A	28-09-1995
		JP 7507803 T	31-08-1995
WO 9508265 A	30-03-1995	AT 167609 T	15-07-1998
		AU 689499 B	02-04-1998
		AU 7638394 A	10-04-1995
		BR 9407709 A	12-02-1997
		CA 2172399 A	30-03-1995
		CN 1131899 A	25-09-1996
		DE 69411276 D	30-07-1998
		DE 69411276 T	26-11-1998
		EP 0720427 A	10-07-1996
		ES 2118433 T	16-09-1998
		HU 76001 A,B	30-06-1997
		JP 9502975 T	25-03-1997
		PL 313733 A	22-07-1996
		TR 27764 A	04-08-1995
		US 5714157 A	03-02-1998
		ZA 9406999 A	12-03-1996
EP 0280289 A	31-08-1988	AT 78658 T	15-08-1992
		DE 3873114 A	03-09-1992
		ES 2042613 T	16-12-1993
		HU 46835 A,B	28-12-1988
		IN 166819 A	21-07-1990
		JP 2706655 B	28-01-1998
		JP 64000004 A	05-01-1989
		KR 9605159 B	22-04-1996
		RU 2027364 C	27-01-1995

This Page Blank (uspto)

**This Page is Inserted by IFW Indexing and Scanning
Operations and is not part of the Official Record**

BEST AVAILABLE IMAGES

Defective images within this document are accurate representations of the original documents submitted by the applicant.

Defects in the images include but are not limited to the items checked:

- ☐ BLACK BORDERS
- ☐ IMAGE CUT OFF AT TOP, BOTTOM OR SIDES
- ☐ FADED TEXT OR DRAWING
- ☒ BLURRED OR ILLEGIBLE TEXT OR DRAWING
- ☐ SKEWED/SLANTED IMAGES
- ☐ COLOR OR BLACK AND WHITE PHOTOGRAPHS
- ☐ GRAY SCALE DOCUMENTS
- ☒ LINES OR MARKS ON ORIGINAL DOCUMENT
- ☐ REFERENCE(S) OR EXHIBIT(S) SUBMITTED ARE POOR QUALITY
- ☐ OTHER: _____

IMAGES ARE BEST AVAILABLE COPY.

As rescanning these documents will not correct the image problems checked, please do not report these problems to the IFW Image Problem Mailbox.

THIS PAGE BLANK (USPTO)